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CHEMISTRY ANDPPHYSICS LAB A B PLUCHINO ET AL

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ABSTRACT

The photophoretic force is described by a comprehensive model containing aspects of both the optical local field and gas dynamic properties. It is shown that the model is in good agreement with detailed experimental data, and provides a new means for measuring optical constants of microparticles.

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Recently there has been a great deal of interest in the local field which is produced by electromagnetic radiation on the surface and within the interior of a microparticle.^{1,2} This interest is due, in part, to various enhancements which are found in photophysical interactions with molecular adsorbates, such as fluorescence³ and Raman scattering.² Since such phenomena are generally observed in the far field, they principally provide information about the surface averaged local field. Photophysical processes which directly measure the distribution of the local field are unknown to us. In this connection, the photophoretic force is of great interest, since its origin lies directly in the anisotropic heating process which is the result of a nonuniform local field distribution. Pluchino⁴ has attempted to fit the size dependence of the radiometric force⁵ by incorporating Mie theory into the current continuum model of Yalamov et al.⁶ He found that although the model was in good qualitative agreement with theory, the measured force was found to be smaller than its calculated counterpart by 100 to 300% depending on size. In what follows we will show that Mie theory can account for the measured shape of the photophoretic force vs. size so long as one is not restrained to continuum hydrodynamics and includes effects associated with the molecular mean-free-path in the external gas. This model is further tested by experimentally examining the dependence of the null point, i.e., the size at which the force on the particle vanishes before reversal, for various complex indices of refraction \hat{n} . Indeed, the results show that null point measurements provide a means for obtaining the imaginary part of \hat{n} , a property not easily measured for particulates by any other technique.

In the limit of low Knudsen number (ratio of molecular mean-free-path to particle radius), a surface temperature gradient ∇T_s causes molecules at the surface to undergo a motion in the direction of the gradient known as

"Maxwellian creep."⁷ At the surface of a sphere, this creep velocity is given by

$$V_{\theta} = \frac{K\eta_e}{RT_s\rho_g} \frac{\partial T_s}{\partial \theta}, \quad (1)$$

where K is the coefficient of thermal slip, T_s is the surface temperature, R is the particle radius, ρ_g is the density of the external gas, η_e is the gas viscosity, and angle θ is defined in Fig. 1. The stress imparted to the gas by the unevenly heated surface produces a reaction on the surface which is the origin of the photophoretic force (photothermal) in the small Knudsen number regime. Yalamov et al.³ have shown by applying such a stress to the surface that the resulting force F_r is

$$\vec{F}_r = - \frac{4\pi R\eta_e^2 J K \vec{I}}{\rho_g T_s K_1}, \quad (2)$$

where K_1 is the interior thermal conductivity, \vec{I} is the incident intensity, and J is a measure of the asymmetry of the internal heat sources. This asymmetry factor is actually a composite of two dimensionless factors, the well known absorption efficiency Q_a and an anisotropy factor A_z , $J = \frac{3}{8} Q_a A_z$.⁵ This anisotropy factor which provides a measure for the distribution of the absorbed energy is given by

$$A_z = \frac{\int_0^1 dx' x'^3 \int_0^\pi d\theta \sin(2\theta) |\vec{E}_1(x', \theta)|^2}{\int_0^1 dx' x'^2 \int_0^\pi d\theta \sin\theta |\vec{E}_1(x', \theta)|^2}, \quad (3)$$

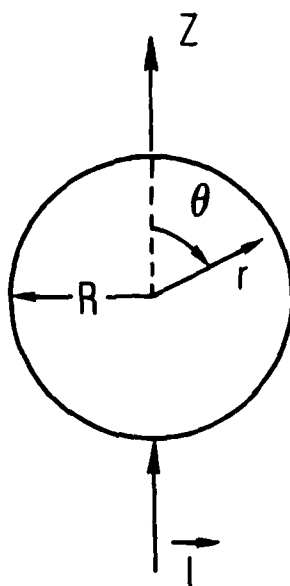


Figure 1. Coordinate system used in evaluating Eqs. (1 and 3).

where x' is the dimensionless length r/R and $|\vec{E}_1(x', \theta)|^2$ is the square modulus of the internal electric field strength averaged over the azimuthal angle ϕ . It is A_2 which controls the sign of the force; with A_2 positive, the back surface is heated most effectively and the particle moves back toward the light source (negative photophoresis). Equation (2) has been applied to the measurement of the photophoretic force on an evaporating glycerol drop. The resulting value of J as reported by Arnold and Lewittes⁵ is shown by the lower curve in Fig. 2. The points in the same figure show the results of a calculation by Pluchino.⁴ As one can see, the agreement is qualitative except for the position of the cross-over point and the sizes corresponding to the apparent resonances in this spectrum. Experimental explanations for this difference such as the use of a Gaussian beam or improper intensity calibration have been ruled out in the interim. This disparity is more fundamental. The model of Yalamov et al⁶ only applies to the case of low Knudsen number, continuum theory. Although the mean-free-path was smaller than 10% of the diameter for the experimental results in Fig. 2, we now look at its possible significance.

Reed⁸ has evaluated the effect on photophoresis of the breakdown of continuum hydrodynamics. However, in this work the particle was assumed to be surface absorbing (i.e., $|E(r, \theta)|^2 = |E(a, \pi)|^2 H(\pi/2 - \theta) \cos \theta$). In either case, Reed⁸ showed that the force should be reduced from the continuum result by a factor

$$g(K_n) = \frac{1}{1+3C_m K_n} \cdot \frac{1}{1+2C_t K_n} \quad (4)$$

where C_m is related to momentum accommodation at the gas-particle interface (a number between 1.00 and 1.35) and C_t is the temperature jump coefficient (a

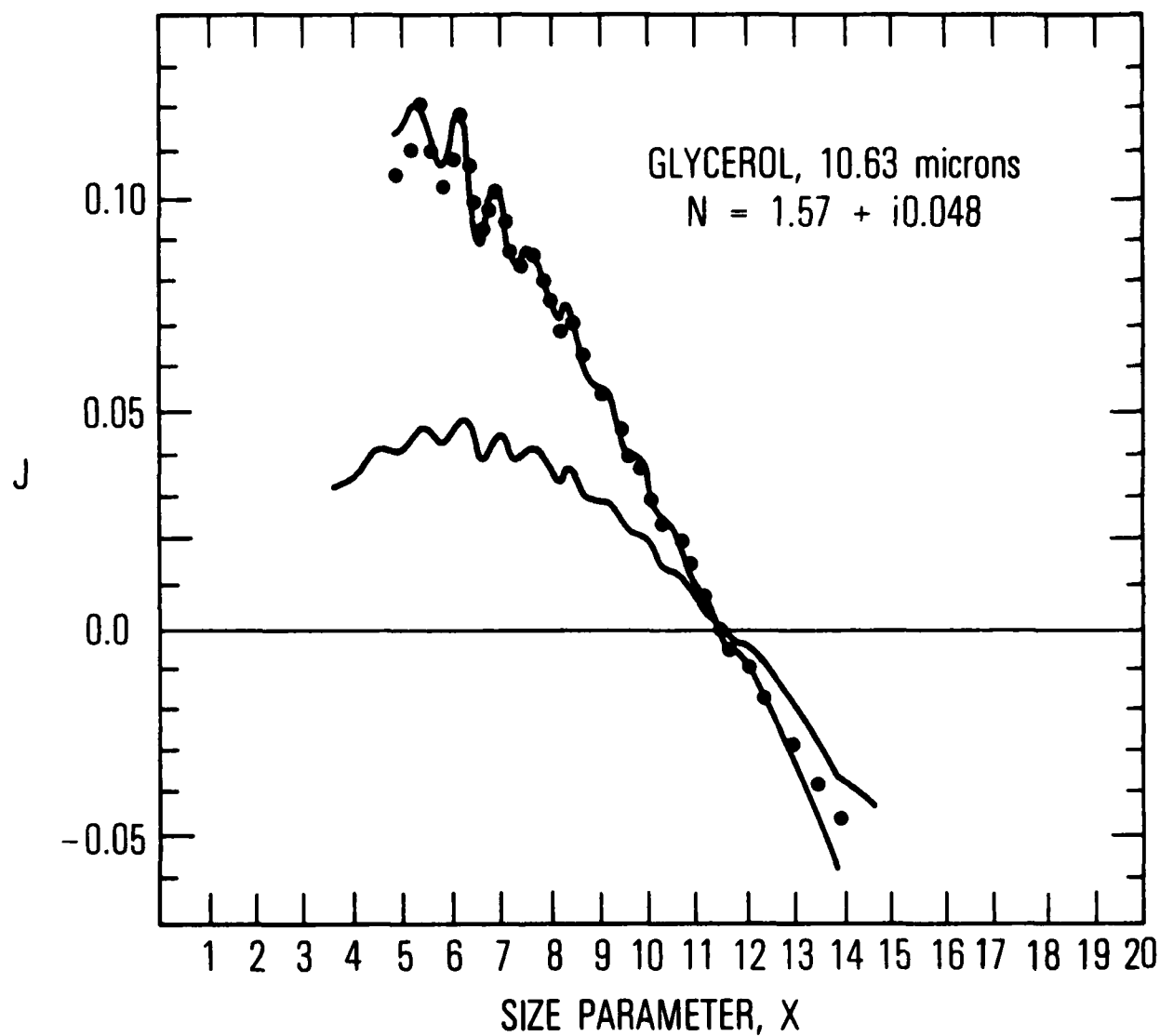


Figure 2. The lowest curve shows experimental values⁵ for J based on Eq. (2). The solid points are a calculation⁴ of J. Top curve presents experimental values for J using the comprehensive model, Eq. (5).

number between 1.875 and 2.48).⁹ In contrast to Reed⁸ our internal electromagnetic field includes all appropriate vector spherical harmonics in the source function, Mie theory. However, this does not preclude the use of Eq. (4). When one considers that only the first Legendre term in the surface temperature distribution drives the force,⁶ the role of the Mie theory is principally one of determining the amplitude of this component. Once having done this we essentially return to Reed's problem. Thus, Eq. (4) will be incorporated as a multiplication factor to the continuum model

$$\vec{F}_{rc} = \vec{F}_r g(K_n) \quad (5)$$

Using typical numbers for C_t and C_m (2.0 and 1.0, respectively) we arrive at the result shown by the top curve in Fig. 2. The agreement is now very good, however, it involves one adjustable constant. The constant is K , Maxwell's so-called "thermal slip coefficient." It is significant that the value of K needed in producing the agreement in Fig. 2 is ~ 1.25 and not $3/4$ as predicted by Maxwell. Some variation in K may be produced by choosing slightly different values of C_t and C_m . However, within the normal range of these constants, K is not reduced by more than 15%. Apparently the disparity between our value and that given by Maxwell requires a more enlightened understanding of the effects of surface scattering on the thermally induced molecular flow field around the particle. Derjaguin and Yalamov¹⁰ have looked at this problem theoretically and find values from 0.75 to 1.5 depending on the degree of diffuseness in the molecular scattering process. Accordingly our value indicates that a significant percentage of scattering events are diffuse. A further test of Eq. (5) which does not depend on either the transport properties in the particle or surrounding gas involves the determination of the "null" point.

Figure 3 shows the experimental setup for null point measurements. The particle is stably trapped in the levitator normally used for photophoretic spectroscopy.¹¹ A disc electrode within this chamber produces a centering force. The particle is sized as in Ref. 5 by producing a shadow image using the forward scattered light from a He-Ne laser. A square wave modulated beam from a tunable CO₂ laser is introduced horizontally causing the particle to move toward or away from the laser source. At large sizes the particle is pushed, as it shrinks, eventually, the force is reversed and the particle is pulled just beyond the null point. This point of reversal is revealed, and visually detected, by a change in phase of 180° of the horizontal fluctuation with respect to the laser modulation.

The results at three distinct wavelengths are shown in Table 1. The refractive indices were obtained from bulk measurements. We see agreement within the uncertainty in size at 10.63 and 10.22 μm . However, the theoretical null point is slightly outside our experimental uncertainty in size at 10.33 μm . If we consider the uncertainty in the measurement of k at this wavelength, a calculated value consistent with the measurement is obtained.

We have shown that photophoresis may be described by a comprehensive model which includes Mie theory and a realistic view of mean-free-path effects. Furthermore, we have demonstrated that the supposition that the imaginary part of the refractive index for a single particle may be measured from the null point,^{4,12} is in fact correct. Data taken in the IR may be expected to be scaled to smaller particles in the visible index. Thus, reversed levitation of a spherical particle 1 μm in diameter, with equivalent visible refractive index, was predicted to occur on the basis of the original model at one solar constant.^{4,5} However, the mean-free-path correction in Eq. (5) considerably reduces the force negating the possibility of spherical

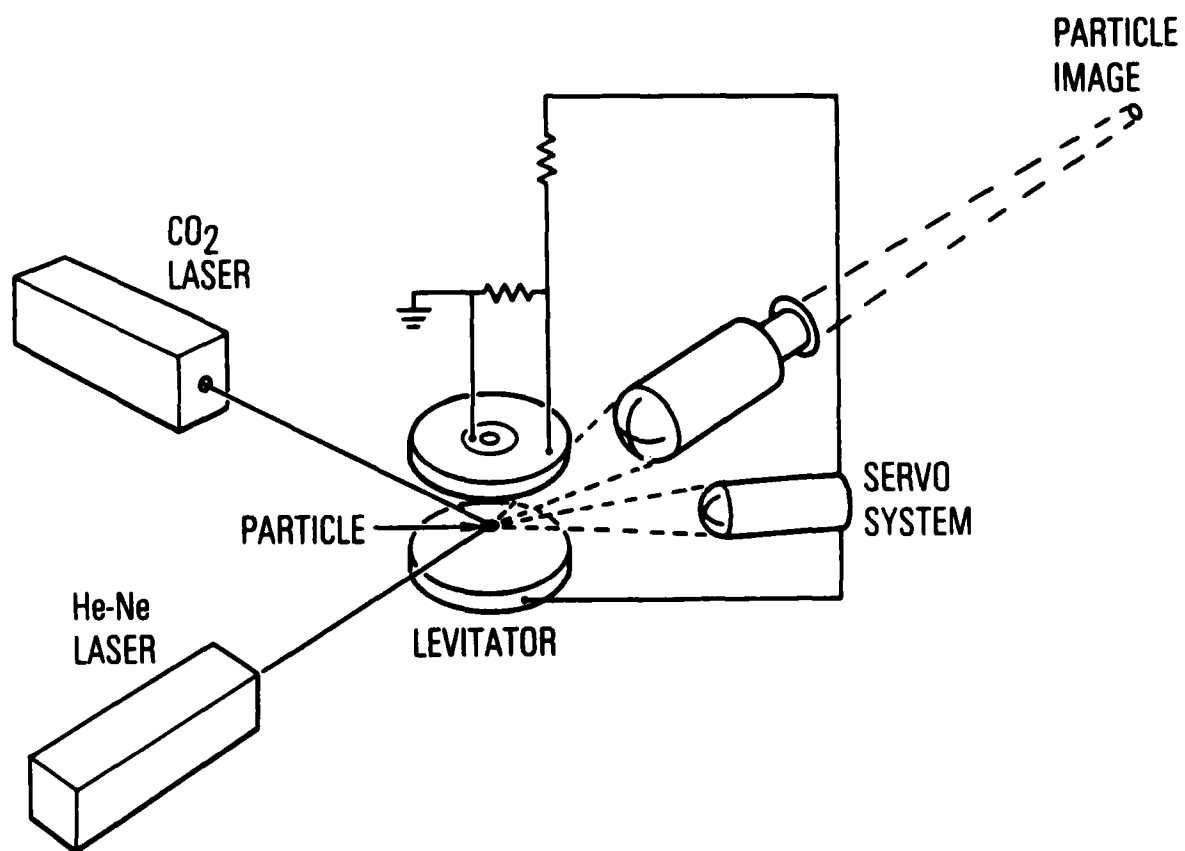


Figure 3. Description of experiment to measure null point.

Table I. Photophoretic Null Point Measurement

$\lambda(\mu\text{m})$	n	EXPERIMENT		THEORY	
		k	x	k	x
10.63	1.55	0.045	11.26 ± 0.5	0.045	11.70
10.33	1.68	0.054	9.61 ± 0.5	0.054	10.50
10.22	1.92	0.095	6.85 ± 0.5	0.095	6.50

particle levitation under terrestrial conditions. In applying our linear model to atmospherical particles such as sulfuric acid droplets, carbon soot, and desert dust, no levitation due to solar irradiation is found. This statement does not include nonspherical particles for which calculations have not been made. Contrary to our findings, Sitarski and Kerker¹³ find that for large Knudsen numbers some of these atmospheric particles will be levitated by solar irradiation.

It is interesting to note that our model has recently been applied to the surface mode region of small particles.¹⁴ We see in this case that photophoresis allows one to detect high order surface modes which are not readily addressed by light scattering. These modes are revealed by an enhancement in A_z .

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